

Modeling and Data Analysis of Aerosol Properties and Radiative Forcing Over the Asian-Pacific Region

PI: *Mian Chin*, Georgia Institute of Technology and NASA Goddard Space Flight Center
chin@rondo.gsfc.nasa.gov

Project Summary

This project applies a global 3-D model to data analysis in the Aerosol Characterization Experiment-Asia (ACE-Asia) mission. The objectives are (1) to understand the physical, chemical, and optical properties of aerosol and the processes that control these properties over the Asian-Pacific region, (2) to determine the aerosol radiative forcing over the Asian-Pacific region, and (3) to assess the influence of anthropogenic activities in Asia on the regional and global atmosphere. The Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model will be used in this investigation. The model is driven by the assimilated meteorological fields from the Goddard Earth Observing System Data Assimilation System (GEOS DAS), with a horizontal resolution of 2° latitude by 2.5° longitude and 20 to 40 vertical levels. The current GOCART model includes the major tropospheric aerosol types of sulfate, carbonaceous, dust, and sea salt. Because it uses assimilated meteorological fields and is a global scale model, the GOCART model can effectively integrate the data from both in-situ and satellite observations, place the properties measured in field campaigns into a broader context, and perform regional and global analyses. A strong link between the model, the in-situ measurements, and the satellite data is the foundation of this project. Approaches are designed as follows: First, all the major types of aerosols will be simulated, and an ammonia cycle will be implemented in the model for estimating sulfate aerosol composition and aqueous phase formation rates. Second, the optical properties of aerosols will be calculated, and an efficient radiative transfer model, consistent with the GEOS DAS fields, will be incorporated into the GOCART model to calculate aerosol radiative forcing. Third, the model calculated physical, chemical, and radiative properties will be evaluated against the in-situ and satellite observations, while a series of sensitivity tests will be conducted to test the parameters used in the model. Finally, the model will be used to analyze and interpret the ACE-Asia data to address the three objectives proposed in this study.

Project Description

1. Introduction

Aerosols affect the climate directly by absorbing or scattering radiation, and indirectly by altering cloud formation and cloud properties. The magnitude of these effects are however poorly constrained, because of our limited knowledge of the processes that control aerosol distributions and the relationships that exist between the aerosols mass and their optical properties. In fact, it has been well recognized that aerosols cause the largest uncertainty in assessing the radiative forcing of climate [IPCC, 1995]. In addition to their role in climate change, aerosols also affect atmospheric chemistry by providing sites for heterogeneous chemistry to take place and to alter the photolysis rates, thus affecting concentrations of many important tropospheric constituents. In turn, changes in the atmospheric chemistry will affect the rates of aerosol formation and growth. Understanding these various effects requires measurements of the physical, chemical, and optical properties of aerosols as well as their spatial and temporal variations.

There have been several multi-platform field missions designed specifically for aerosol process and property studies. These missions have been conducted in key areas of the globe. For example, the first Aerosol Characterization Experiment (ACE-1, November - December 1995) studied natural, unpolluted aerosols in the remote southern ocean in Nov-Dec 1995 [Bates et al., 1998]. The Tropospheric Aerosol Radiative Forcing Experiment (TARFOX, July 1996) focused on the radiative effect of anthropogenic aerosols off the east coast of North America [Russell et al., 1999]. ACE-2 (June - July 1997) targeted at European pollution and African dust aerosols and their impact on the North Atlantic [Raes et al., 1999]. More recently, the Indian Ocean Experiment (INDOEX, January - March 1999) was aimed to understand the radiative forcing of the anthropogenic and natural aerosols from the southeastern Asia, southern Africa, and from the ocean. These field programs have provided valuable information on the chemical, physical, and radiative properties of aerosols at different regions of the globe.

Now the ACE-Asia experiment is planned to take place in the Asian-Pacific region in the period of 2000 to 2004. The uniqueness of aerosol in the Asian-Pacific region, in contrast with that in Europe and North America, has several aspects. (1) Sources. East Asia is an important source region of all major tropospheric aerosols. The fast economic development, the large area of desert, and the intensive forest and agriculture fires in this region contribute to 1/4 to 1/3 of global emissions of SO₂, organic matter, soot, and dust [Chin et al., 1999; Ginoux et al, 2000; Liousse et al., 1996]. (2) Properties. Asian aerosols are probably always a mixture of multiple components. For example, mineral dust transported out from the desert can pass major pollution regions where it can be coated and mixed with sulfate and organic aerosols. It can be further mixed with sea salt particles off the coast. The physical, chemical, and radiative properties of the multi-

component mixture are more complex than any single type of aerosols. To reduce the overall uncertainty in the calculation of aerosol climate forcing and to understand the multiphase atmospheric chemical system, ACE-Asia will pursue three specific objectives: (1) to determine the properties of the major aerosol types; (2) to quantify the processes controlling those properties; and (3) to extrapolate the properties and processes from local to regional and global scales, and assess the radiative forcing by aerosols in the Asian-Pacific region (ACE-Asia Prospectus, <http://sage.peml.noaa.gov/aceasia>).

In accord with the ACE-Asia objectives, we propose here a 3-year research project using the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model to combine satellite and ACE-Asia data for aerosol analysis. The GOCART model is driven by the assimilated meteorological fields from the Goddard Earth Observing System Data Assimilation System (GEOS DAS), therefore it is suitable for use in the field campaigns. On the other hand, it is a global model so it can be used to effectively integrate the field and satellite observations and to place the ACE-Asia data (and other in-situ data) into a global context. This proposed research is an enhancement of and complimentary to the PI's other ongoing projects of tropospheric aerosols, supported by NASA and NOAA. These projects involve developing a global 3-D aerosol model and using it to analyze the aerosol data from in-situ measurements as well as from satellite observations.

In the following sections we will briefly describe the GOCART model and present some results (section 2), and then we will state our objectives (section 3) and approach (section 4) for this project. Collaborations with other investigations and computational resources will also be listed (sections 5 and 6).

2. The GOCART model

The GOCART model used in this project is a global scale model driven by the GEOS DAS assimilated meteorological fields. It has a horizontal resolution of 2° latitude by 2.5° longitude and 20 to 40 vertical layers (vertical resolution depends on the version of GEOS DAS; see below). The GEOS DAS fields are model-assimilated global analyses constrained by meteorological observations, with extensive diagnostics archived for chemistry transport model applications [Schubert et al., 1993]. There are so far 3 versions of the GEOS DAS generated assimilated meteorological data, each covering a particular time period with a different vertical resolution. GEOS-1 data are in 20 sigma layers with a model top pressure of 10 mb; they are available for the period from January 1980 to February 1995. GEOS-1.3 (also known as GEOS-STRAT) has 46 vertical layers, 26 to 29 of which are in the stratosphere and the model top is 0.1 mb. GEOS-1.3 data are available for the period from February 1995 to early 1998. From December 1998, the newest version GEOS-2 (or GEOS-TRMM) takes over. GEOS-2 has a much higher model top of 0.003 mb with 70 vertical layers; only 27 to 30 of these layers are in the troposphere. For tropospheric applications and computational efficiency, we have

aggregated the GEOS-1.3 top 23 layers into 3 such that the total vertical resolution is 26 layers, with 6 to 9 in the stratosphere. We plan to aggregate the GEOS-2 data into a total of 40 layers, with 10 to 13 of them in the stratosphere.

The current GOCART model has the capability of simulating major components of tropospheric aerosols, including sulfate, dust, sea salt, and carbonaceous aerosols. The model contains the following modules in aerosol simulation: emission, which includes sulfur, dust, black carbon and organic carbon, and sea salt emissions; chemistry, which currently uses prescribed OH, H₂O₂, and NO₃ fields for gaseous sulfur oxidations; advection, which is computed by a flux-form semi-Lagrangian method [Lin and Rood, 1996]; boundary layer turbulent mixing, which uses a second-order closure scheme [Helfand and Labraga, 1988]; moist convection, which is calculated using archived cloud mass flux fields [Allen et al., 1996]; dry deposition, which uses a resistance-in-series algorithm [Wesely, 1989] as a function of surface type and meteorological conditions; and wet deposition, which accounts for the scavenging of soluble species in convective updrafts and rainout/washout in large-scale precipitation [Balkanski et al., 1993]. We consider 4 size bins for dust ($r=0.1-1$, $1-1.8$, $1.8-3$, $3-6$ μm) as well as for sea salt ($r=0.1-0.5$, $0.5-1.5$, $1.5-5$, $5-10$ μm). For sulfate and carbonaceous aerosols we have been calculating their mass without resolving their sizes.

We have evaluated the modeled sulfate (and its precursors) with extensive long term data from surface networks at polluted regions of North America and Europe and over the global oceans. We have also compared model results with aircraft and ship observations in several field campaigns [Chin et al., 1998, 1999, 2000]. Some of the results are shown in Figure 1. Particularly relevant to ACE-Asia are the results at Cheju, Okinawa, Midway, Oahu, and Mauna Loa (Figure 1b), where the model captures the spring time transport of Asian anthropogenic sulfate along the pathway to reach Hawaii. The model also reproduces the vertical profiles taken from the aircraft during the NASA Pacific Exploratory Mission (PEM)-West B, a mission designed to measure the Asian outflow in the spring (Figure 1c). Modeled dust sources and transport have also shown an excellent match with the satellite observations (Figure 2) [Ginoux et al., 2000]. Recently, participating in the IPCC assessment report of aerosol radiative forcing, we have incorporated carbonaceous and sea salt aerosols into the model. The total aerosol optical thicknesses have been compared with those obtained from the ground-based sun photometer measurements in the Aerosol Robotic Network (AERONET) stations and those derived from TOMS satellite retrievals (Figure 3).

3. Objectives

(1) Understand the chemical, physical, and optical properties of aerosols and the processes controlling these properties over the Asian-Pacific region.

The uniqueness of aerosols over the Asian-Pacific region is that it is composed by several types from both natural and anthropogenic sources. Table 1 lists the major

sources of sulfate, carbonaceous, dust, and sea salt in the globe and in the Asian-Pacific region (latitude 0° - 60°N, longitude 60° - 180°E) used in our recent studies. The greatest source strengths are sea salt and dust. However, because these aerosols are usually in large size, the regional distributions of aerosol optical thickness are not necessarily dominated by these two aerosol populations. Figure 4 shows the preliminary GOCART model results of total aerosol optical thickness at 550 nm, compared with 630 nm quantity retrieved from the satellite sensor AVHRR (Advanced Very High Resolution Radiometer). It seems that, on annual average, dust contributes the most to the total optical thickness in the region, although over the ocean it is comparable to sulfate. Figure 4 also shows that organic carbon is important in the eastern and southern part of the east Asian continent, while dust dominates the northern and western part. The optical thickness of sea salt is small even over the ocean. A major difficulty however in converting model calculated aerosol mass to the optical thickness is the large uncertainties in estimating the extinction coefficients for different types of aerosols in different environment.

Table 1. Source strength (Tg/year) for major aerosols from the GOCART model.

Type	Asian-Pacific	Global	Reference
Sulfate (emission + conversion)	105	360	Chin et al., 1999
Black Carbon (emission)	4	12	IPCC 2000 ¹
Organic Carbon (emission + conversion)	23	83	IPCC 2000 ¹
Dust (emission)	593	2074	Ginoux et al., 2000
Sea Salt (emission)	582	5823	Ginoux, unpublished

¹IPCC emission scenario for the present day (provided by J. Penner, 1999).

Aerosols may also form complex mixtures, both externally mixed (individual particles having separate identities) and internally mixed (more than one type of aerosols mixed within a single particle). Optical properties for internally mixed aerosols are not equivalent to those of an external mixture with the same composition. For example, the less soluble organic species mixed with sulfate will increase light absorption and decrease the solubility of sulfate particles. Similarly, sulfate or sea salt mixed with dust will reduce the absorption and increase the apparent solubility of dust aerosols. In addition to dry deposition on the surface and the wet removal, aerosols can collide to each other to increase their removal rate (e.g., nitrate aerosol loss on dust and biomass burning particles, Tabazadah et al., 1998). The state of mixing is particularly important in the Asian-Pacific region, since the aerosol over that region is almost always a mixture of multiple components with dust, sulfate, carbonaceous, and sea salt. Niimura et al. [1998] found that from 16 to 100 % of Asian dust storm particles were internally mixed with sea salt after they passed the Yellow Sea and reached Nagasaki in Japan. The state of mixing and thus the hygroscopic and optical properties of the multi-component Asian aerosols is expected to be more complex than aerosols from other regions of the world.

Different types of aerosols or their precursor gases can also chemically interact with each other. A number of studies have shown that the presence of sea salt and mineral dust aerosols should be accounted for the loss of gas-phase SO_2 , which is a major precursor for sulfate aerosol. For example, Sievering et al. [1999] estimated from the ACE-1 measurements that SO_2 oxidation on sea salt aerosol could account for 30-35% of the non-sea salt sulfate formation. Dentener et al. [1996] suggested that the reaction of SO_2 with calcium-rich mineral dust plays an important role in SO_2 conversion downwind of arid source regions, such as Asia. Model calculations by Xiao et al. [1997] suggested that in the Asian-Pacific region chemical conversion of SO_2 to sulfate in the presence of mineral aerosol may contribute 20-40% of total sulfate production rate in early spring when gas-phase reactions are comparatively slow. With the co-existence of sea salt, mineral dust, and anthropogenic pollution aerosols, the ACE-Asia study offers the unique opportunity to observe the interactions between these components.

(2) Determine the aerosol radiative forcing over the Asian-Pacific region

The indirect effect of aerosol on climate forcing is generally thought to be the most uncertain since there is only a limited knowledge of the complexity of cloud formation mechanisms; we know as little about the ways aerosols affect these mechanisms. Although the direct effect of aerosols is considered to be less uncertain, the sign of this forcing is still very much in question because it depends on the aerosol composition, vertical profiles, and size distribution. Sulfate aerosol is considered to have pure scattering effect, thus causing cooling, while dust and black carbon absorbs heat and may cause warming.

Modeling of direct aerosol radiative forcing requires the optical thickness, single scattering albedo, and a phase function P (a function of wavelength, refractive index, and size distribution) which is often replaced by a simplified backscatter coefficient. Since most aerosols absorb water at higher relative humidities, their size and scattering efficiency vary with relative humidity. This variation is however strongly dependant on aerosol types. Given the complexity of the mixing state and thus the hygroscopic and optical properties of the Asian aerosols, the parameters used previously in the model for calculating radiative properties may not be appropriate. Measurements in ACE-Asia will help to improve the parameterization of aerosol properties.

(3) Assess the global impact of anthropogenic activities in East Asia

East Asia is one of the most rapidly developing regions in the world. The fast growth of industry and rapid increase of the number of automobiles have caused and will continue to cause the increase of anthropogenic emissions. These emissions will no doubt change the aerosol composition and concentrations. The export of anthropogenic aerosols and their precursors modifies the atmosphere over not only the western Pacific, but also the whole Pacific basin and even North America. The eastern Pacific atmosphere has been one of the cleanest on Earth because of the size of the ocean, the active wet removal of soluble species, and the relatively low input of aerosols and their precursors.

However, based on the analysis of PEM and ACE-1, Thornton et al. [1999] concluded that anthropogenic sources from East Asia are now dominating the sulfur chemistry in large region of the Pacific atmosphere. Studying the changes caused by the East Asian industrial development will help understand the sensitivity of aerosol formation and removal to the emission change.

To identify the Asian outflow pathways and its characterization, it is critical to combine in-situ data with satellite observations in model analysis. In-situ data have detailed chemical and physical information but are limited in time and space coverage. By contrast, satellite data provide extensive time and space coverage but offers little information on aerosol composition and profile. Successful linking of in-situ measurements, satellite data, and 3-D model products will allow us to extract detailed aerosol information on an extended time and spatial scale, and place the ACE-Asia data into a broader context.

4. Research Plan

A research plan for a 3-year period of 2000 - 2003 is presented here. This period overlaps with the ACE-Asia ground network operations (2000 - 2004) and covers intensive operations planned for the spring of 2001. We intend to continue our investigation beyond the 3-year period in coordination with ACE-Asia intensive campaign period for cloud-aerosol interaction study. Table 2 shows the timeline of this investigation along with the ACE-Asia measurement plan.

Table 2. Timeline for this project (3-year) as well as the ACE-Asia measurement plan.

	2000	2001	2002	2003	2004
ACE-Asia measurements:					
Ground network	<----->				
Phys/Chem/Rad. intensive		<----->			
Cloud-Aerosol interaction intensive			<-----> or	<----->	
Research Plan:					
Aerosol and simulations	<----->				
Model evaluation	<----->				
Analysis of ACE-Asia data		<----->			

4.1. Simulation of major tropospheric aerosols

Based on our current aerosol modeling capability with the GOCART, we will continue to improve the simulations of major tropospheric aerosol types. In our current simulations we calculate each aerosol type separately. In this investigation, we will simulate all aerosol types together in order to test the degree of interactions between different types of aerosol. For example, we will test the loss rate of SO₂ on sea salt or dust, and examine how the mixing state of sulfate/dust/carbonaceous/sea salt affects the optical properties and removal processes of aerosols.

We will add the ammonia cycle to the model to calculate the pH values of cloud and rain droplets and the composition of sulfate aerosol. Ammonia plays a key role in determining the pH of the cloud and rain droplets, because it is able to neutralize a great portion of sulfuric and nitric acids. The droplets pH values will affect the aqueous phase reaction rates of SO_2 with H_2O_2 and ozone. The composition of sulfate aerosol is also important in radiative property calculations, since the deliquescence properties of sulfate aerosol depend on the amount of ammonium present. We will, based on the sulfur modules, incorporate the global ammonia emission data inventory [Bouwman et al., 1997] and reactions of ammonia with sulfuric acid, NO_x , and HO_x into the GOCART model.

4.2. Calculation of aerosol direct radiative forcing

With a full range of aerosol simulated, we will be able to calculate the aerosol single scattering albedo and optical thickness. We will also improve the model parameterization of the humidification effects on aerosol size and scattering coefficient. Currently, the model includes a number of discrete size bins of dust and sea salt aerosols, and we plan to assume a size distribution for sulfate and carbonaceous aerosols. We will also explore the feasibility of using a microphysics model of Toon et al. [1988] in conjunction with the GOCART for size-resolved aerosol simulation. The aerosol optical thickness, size distribution, single scattering albedo, and scattering efficiency can then be used to calculate the direct forcing of aerosol. A radiative transfer model [Chou et al., 1992] will be incorporated into the GOCART model. Absorption by gas species, scattering by air molecules, and absorption and scattering by different types of aerosol will be calculated over a wide range of spectrum (0.175 μm to 10 μm). The same radiative transfer model is also used in the GEOS DAS model analysis system, so which should be consistent with the meteorological data used in the GOCART model.

4.3. Model evaluation with satellite, ground-based network, and in-situ field data

The model will be evaluated with data from satellite, ground-based network, and field campaigns. We will compare the model generated spatial distribution patterns with satellite data, concentrations with field and ground-based data, and optical thicknesses with sun photometer and satellite data. The field data for model evaluation include ACE-1, ACE-2, TARFOX, and INDOEX. We plan to use the aerosol products from the two best known satellite sensors: AVHRR and TOMS (Total Ozone Mapping Spectrometer). AVHRR reports aerosol optical thickness at visible wavelengths over the ocean [Husar et al., 1997], and TOMS provides aerosol index (a qualitative or semi-quantitative measure of aerosol amount) and aerosol optical thickness at UV wavelengths over both land and ocean [Herman et al., 1997; Torres et al., 1998]. With the launch of EOS Terra satellite last December, we should be able to obtain better aerosol data from the sensors MODIS, MISR, and CERES.

Table 3 lists the data sets and measured quantities we will use for model evaluation and later for data analysis. We will then identify the strengths and weaknesses of the model to make improvements.

Table 3. Satellite and field aerosol measurements for model evaluation and data analysis.

	Time	Measured area	Main Products	Notes
<i>Satellite measurements:</i>				
TOMS	since 1978	Global	Absorbing and scattering aerosol optical thickness	Available over both land and ocean
AVHRR	since 1979	Global	Aerosol optical thickness	Available only over the ocean
MODIS, MISR, CERES	after Nov, 1999	Global	Aerosol, cloud, water vapor, vegetation map, radiative budget	Over land and ocean, information on size and composition
<i>Ground-based network measurements:</i>				
AERONET	since 1993	Stations over global land	Aerosol column optical thickness	Nearly 200 stations, many of them new
U. Miami	since 1980's	Ocean and land	Sulfate, nitrate, MSA, dust, sea salt, ^{210}Pb	Surface air concentrations at nearly 30 stations, most of them on islands
<i>In-situ field measurements:</i>				
ACE-1	Nov.-Dec., 1995	Near Cape Grim, Australia	Major aerosol types, particle size, optical properties, gas species	Aerosol in the minimally polluted marine troposphere
TARFOX	July, 1996	Western North Atlantic, near Wallops Island	Major aerosol types, particle size, optical properties, gas species	Anthropogenic aerosols from North America
ACE-2	June-July, 1997	Eastern North Atlantic	Major aerosol types, particle size, optical properties, gas species	Anthropogenic aerosols from Europe; dust from Africa
INDOEX	Jan.-Mar, 1999	Indian Ocean	Major aerosol types, particle size, optical properties, etc.	Aerosols from Asia and Africa, biogenic and sea salt aerosols from ocean
ACE-Asia	2000-2004	Western Pacific	Major aerosol types, particle size, optical properties, gas species	Anthropogenic and dust aerosols from Asia, biogenic and sea salt aerosols from ocean

4.4. Linking in-situ and Satellite Observations to Analyze and Interpret ACE-Asia Data

Based on satellite and ACE-Asia data, we will use the model to identify and quantify the amount, composition, characteristics, and the evolution of aerosols in the outflow from the Asian continent to the Pacific. Although the ACE-Asia field intensive studies are planned for the spring seasons when the Asian export to the Pacific is expected to be at the maximum, it is necessary to analyze other seasons as well to

estimate the anthropogenic influence from other regions, for example, long-range transport from Europe and North America.

Satellite data can provide synoptic coverage of aerosol fluctuations. This is particularly useful for ACE-Asia study, for example, to monitor the dust outbreak episodes, such as the one occurred in spring 1998. As shown in Figure 5, this dust episode is clearly shown in TOMS aerosol index. Large scale aerosol map can also be used to reconcile the model and the in-situ data. We will utilize the daily satellite coverage to help the model interpret the ACE-Asia field observations in terms of aerosol source strength, transport pattern, and the impact of Asian outflow on the the Pacific Ocean.

5. Collaborative Research

This project will be closely coordinated with other on-going or planned projects we have been involved in. Mian Chin is the PI of the project, supported by NASA Global Aerosol Climate Program (GACP), on global model analyses of the retrospective satellite aerosol data. Mian Chin is a Co-I in a NASA EOS/IDS project (China Metro-Agro Plex or China-MAP, PI: W. Chameides, Georgia Tech) for modeling the aerosol and ozone measurements in China and the pollution export from China. Both projects are highly relevant and complementary to the present proposal: the GACP project supports the aerosol model development and satellite data analysis, and the China-MAP project will allow close collaborations with scientists in China to gain better assessment of atmospheric responses to the economic development in China.

We will work closely work with the Goddard TOMS aerosol project team in using and analyzing the TOMS aerosol products. We will investigate the interactions between aerosols and tropospheric chemistry in collaboration with Lyatt Jaegle at University of Washington, who will use the Harvard/GEOS model, which is driven by the same meteorological fields as used in the GOCART model, to simulate important chemical species including ozone, NO_x , HO_x in the ACE-Asia context. We will share our model results and conduct studies on, for example, the effect of aerosols on photolysis rate, the change of aerosol formation and concentration in response to the change of oxidants concentrations, and the effect of aerosols on ozone production rates.

6. Computational Resources

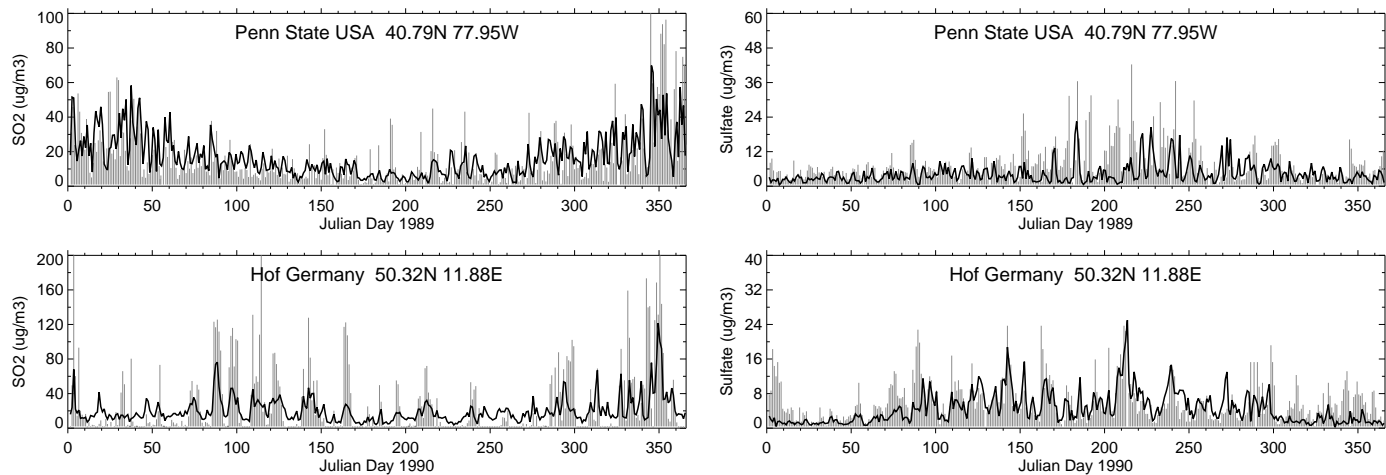
The GOCART model simulations have been running on Cray J90 machine at the NASA Center for Computational Sciences (NCCS) located at GSFC. The GEOS DAS fields and the model output are stored in the NCCS mass storage space. The model is vectorized and parallelized for multi-tasking. The 20-layer version of the aerosol model (4 sulfur species with off-line chemistry, 3 carbonaceous species, 4 dust sizes, and 4 sea-salt sizes) requires about 8 hours real time per model month with 4 processors. There are also several SGI workstations available for data analysis.

References

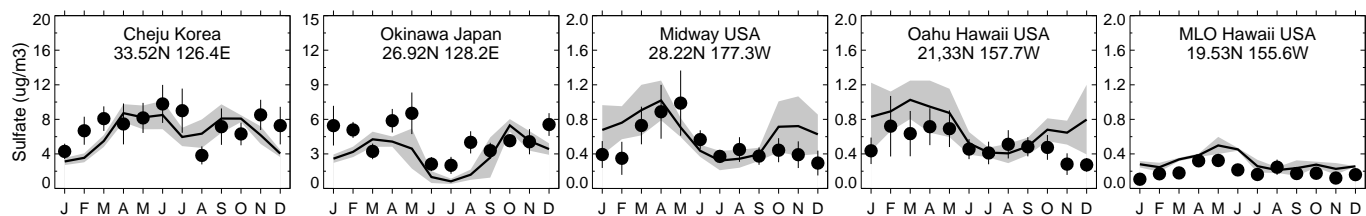
- Allen, D. J., P. Kasibhatla, A. M. Thompson, R. B. Rood, B. G. Doddridge, K. E. Pickering, R. D. Hudson, and S.-J. Lin, Transport-induced interannual variability of carbon monoxide determined using a chemistry and transport model, *J. Geophys. Res.*, **101**, 28,655-28,669, 1996.
- Balkanski, Y. J., D. J. Jacob, G. M. Gardner, W. C. Graustein, and K. K. Turekian, Transport and residence times of tropospheric aerosols inferred from a global three-dimensional simulation of ^{210}Pb , *J. Geophys. Res.*, **98**, 20,573-20,586, 1993.
- Bates, T. S., B. J. Huebert, J. L. Gras, B. Griffiths, and P. A. Durkee, The International Global Atmospheric Chemistry (IGAC) Project's First Aerosol Characterization Experiment (ACE-1) - Overview, *J. Geophys. Res.*, **103**, 16,297-16,318, 1998.
- Bouwman, A. F., D. S. Lee, W. A. H. Asman, F. J. Dentener, K. W. Van Der Hoek, and J. G. J. Olivier, A global high-resolution emission inventory for ammonia, *Global Biogeochem. Cycles*, **11**, 561-587, 1997.
- Chameides, W. L., and A. W. Stelson, Aqueous-phase chemical processes in deliquescent sea-salt aerosols: A mechanism that couples the atmospheric cycles of S and sea salt, *J. Geophys. Res.*, **97**, 20,565-20,580, 1992.
- Chin, M., R. B. Rood, D. J. Allen, M. O. Andreae, A. M. Thompson, S.-J. Lin, R. M. Atlas, and J. V. Ardizzone, Processes controlling dimethyl sulfide over the ocean: Case studies using a 3-D model driven by assimilated meteorological fields, *J. Geophys. Res.* **103**, 8341-8353, 1998.
- Chin, M., R. B. Rood, S.-J. Lin, D. J. Jacob, J.-F. Muller, and A. M. Thompson, Atmospheric sulfur cycle in the global model GOCART: Model description and global properties, submitted to *J. Geophys. Res.*, 1999.
- Chin, M., D. L. Savoie, B. J. Huebert, A. R. Bandy, D. C. Thornton, T. S. Bates, P. K. Quinn, E. S. Saltzman, and W. J. De Bruyn, Atmospheric sulfur cycle in the global model GOCART: Comparison with observations and regional budgets, submitted to *J. Geophys. Res.*, 2000.
- Chou, M. D., A solar radiation model for use in climate studies, *J. Atmos. Sci.*, **49**, 762-772, 1992.
- Dentener, F. J., G. R. Carmichael, Y. Zhang, J. Lelieveld, and P. J. Crutzen, Role of mineral aerosol as a reactive surface in the global troposphere, *J. Geophys. Res.*, **101**, 22,869-22,889, 1996.
- Ginoux, P., M. Chin, I. Tegen, D. Savoie, J. Prospero, B. Holben, and S.-J. Lin, Global simulation of tropospheric dust: 1. Model description and evaluation, to be submitted to *J. Geophys. Res.*, 2000.
- Helfand, H. M., and J. C. Labraga, Design of a nonsingular level 2.5 second-order closure model for the prediction of atmospheric turbulence, *J. Atmos. Sci.*, **45**, 113-132, 1988.
- Herman, J. R., P. K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier, Global distribution of UV-absorbing aerosols from Nimbus-7/TOMS data, *J. Geophys. Res.*, **102**, 16,911-16,921, 1997.
- Husar, R. B., J. M. Prospero, and L. L. Stowe, Characterization of tropospheric aerosols over the oceans with the NOAA advanced very high resolution radiometer optical thickness product, *J. Geophys. Res.* **102**, 16,889-16,909, 1997.
- Intergovernmental Panel on Climate Change, *Climate Change 1994: Radiative Forcing of Climate Change*, edited by J. T. Houghton et al., Cambridge University Press, 1995.
- Kaufman, R. J., D. D. Herring, K. J. Ranson, and G. J. Collatz, Earth Observing System AM1 mission to earth, *IEEE Trans. Geoscience and Remote Sensing*, **36**, 1045-1055, 1998.

- Lin, S.-J., and R. B. Rood, Multidimensional flux-form semi-Lagrangian transport schemes, *Mon. Weather Rev.*, 124, 2046-2070, 1996.
- Liousse, C., J. E. Penner, C. Chuang, J. J. Walton, H. Eddleman, and H. Cachier, A global three-dimensional model study of carbonaceous aerosols, *J. Geophys. Res.* 101, 19,411-19,432, 1996.
- Niimura, N., K. Okada, X.-B. Fan, K. Kai, K. Arao, G.-Y. Shi, and S. Takahashi, Formation of Asian dust-storm particles mixed internally with sea salt in the atmosphere, *J. Meteor. Soc. Japan*, 76, 275-288, 1998.
- Raes, F., T. S. Bates, F. McGovern, and M. Vanlieckekerke, The second Aerosol Characterization Experiment (ACE-2): Overview, *Tellus*, in press, 1999.
- Russell, P. B., P. V. Hobbs, and L. L. Stowe, Aerosol properties and radiative effects in the United States east coast haze plume: An overview of the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX), *J. Geophys. Res.* 104, 2213-2222, 1999.
- Schubert, S. D., R. B. Rood, and J. Pfaendner, An assimilated data set for earth science applications, *Bull. Amer. Meteorol. Soc.*, 74, 2331-2342, 1993.
- Sievering, H., J. Boatman, E. Gorman, Y. Kim, L. Lundersen, G. Ennis, M. Luria, and S. Pandis., Removal of sulfur from the marine boundary layer by ozone oxidation in sea-salt aerosols, *Nature*, 360, 571-574, 1992.
- Tabazadeh, A., et al., Nitric acid scavenging by mineral and biomass burning aerosols, *Geophys. Res. Lett.*, 25, 4185, 1998.
- Toon, O. B., R. P. Turco, D. Westphal, R. Malone, and M. S. Liu, A multi-dimensional model for aerosols: Description of computational analogs, *J. Atmos. Sci.*, 45, 2123-2143, 1988.
- Torres, O., P. K. Bhartia, J. R. Herman, Z. Ahmad, and J. Gleason, Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation. Theoretical bases, *J. Geophys. Res.* 103, 17,099-17,110, 1998.
- Thornton, D. C., A. R. Bandy, B. W. Blomquist, A. R. Driedger, and T. P. Wade, Sulfur dioxide distributions over the Pacific Ocean 1991-1996, *J. Geophys. Res.*, 104, 5845-5854, 1999.
- Wesely, M. L., Parameterization of surface resistance to gaseous dry deposition in regional-scale numeric models, *Atmos. Environ.*, 23, 1293-1304, 1989.
- Xiao, H., G. R. Carmichael, and J. Durchenwald, Long-range transport of SO_x and dust in East Asia during the PEM B experiment, *J. Geophys. Res.*, 102, 28,589-28,612, 1997.

(a) Daily SO₂ and Sulfate over N.America and Europe. Black line: model. Grey vertical bars: obs.



(b) Sulfate Seasonal Cycle Over the Pacific Ocean. Line: model. Circles: obs.



(c) SO₂ and Sulfate vertical profiles in PEM-West B, Feb-Mar 1994. x: model, o: obs.

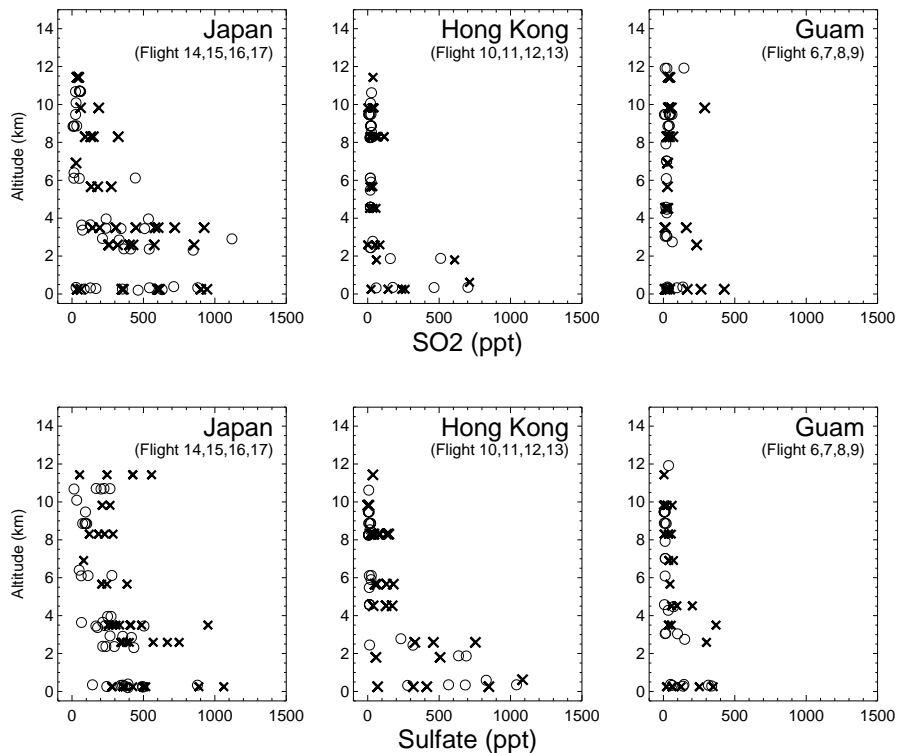
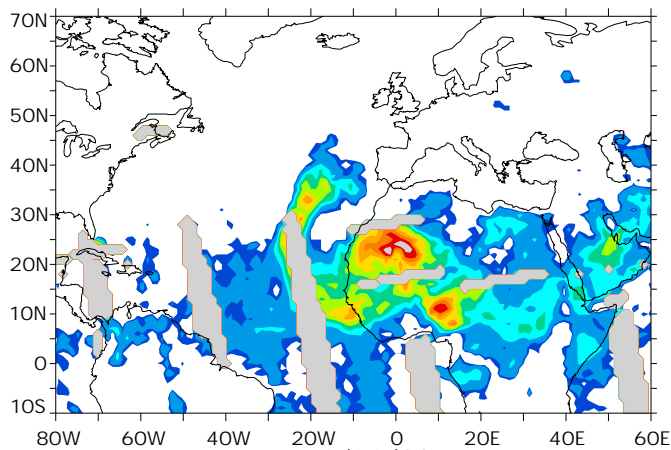


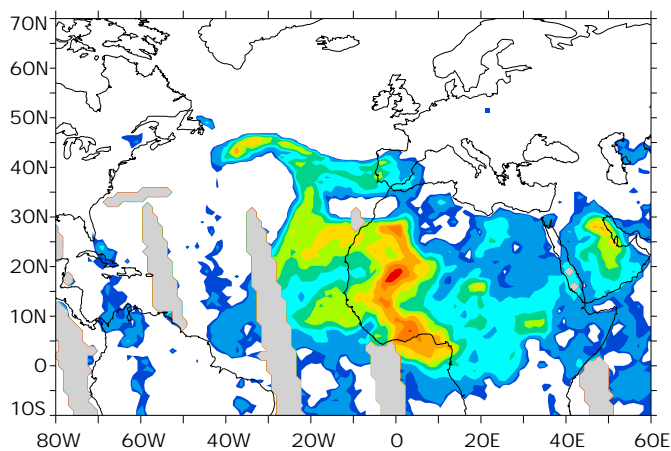
Figure 1. Comparison of model results with observations of SO₂ and sulfate

TOMS Absorbing Aerosol Index

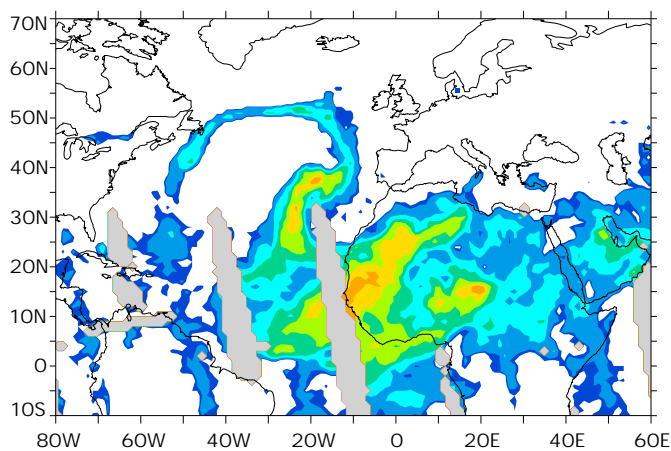
03/27/88



03/29/88

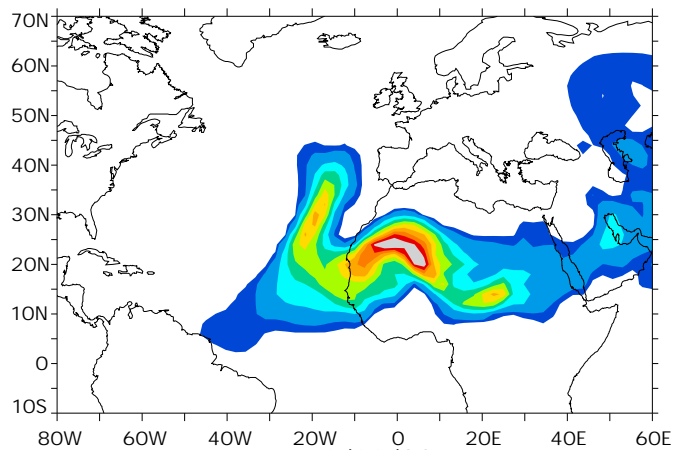


03/31/88

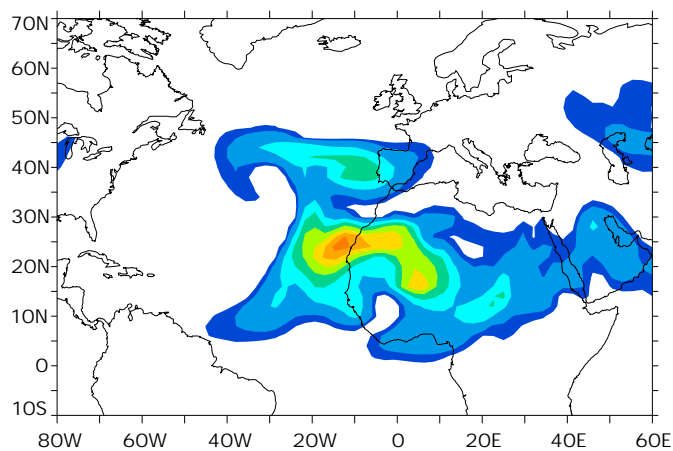


GOCART Dust Mass Column

03/27/88



03/29/88



03/31/88

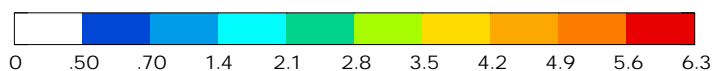
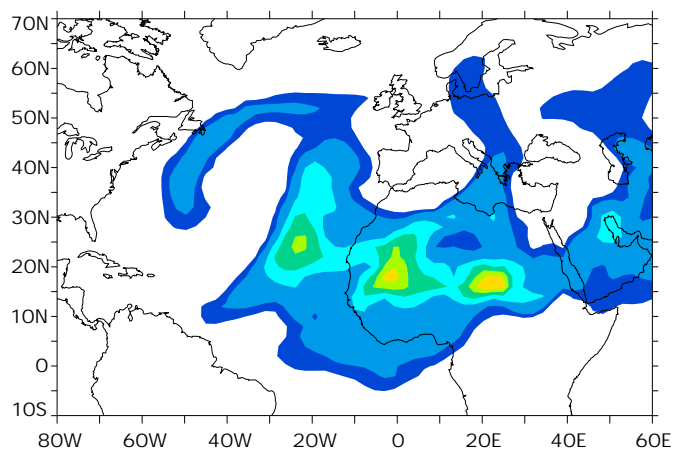


Figure 2. Comparison of model results with TOMS aerosol index for March 1988. Left panels: TOMS index. Right panels: model dust column in 0.4g/m^2 .

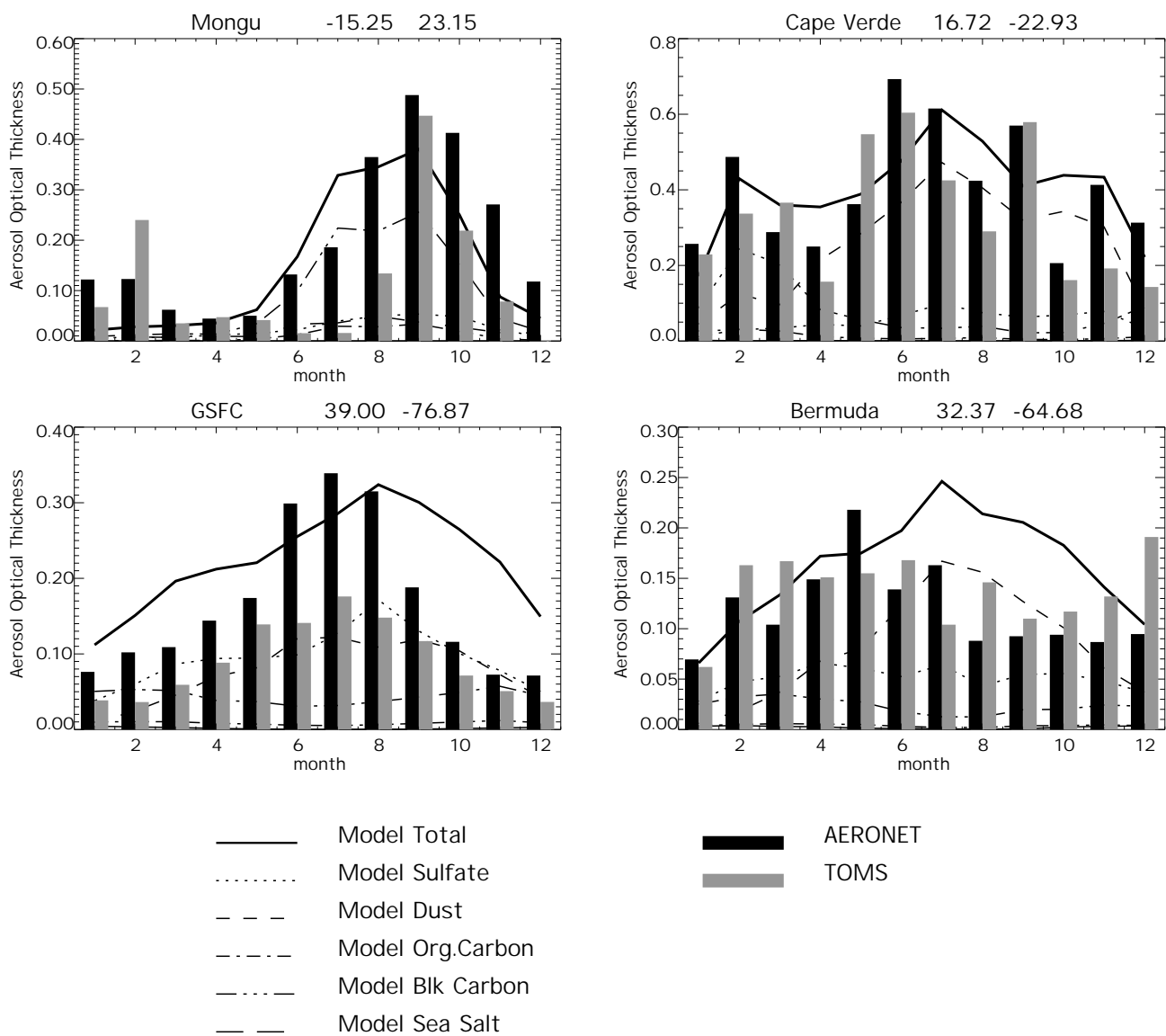


Figure 3. Comparison of monthly avg optical thickness from the model with AERONET and TOMS data at 550 nm. Data are provided by Stefan Kinne and Omar Torres, GSFC.

Annual Avg Aerosol Optical Thickness

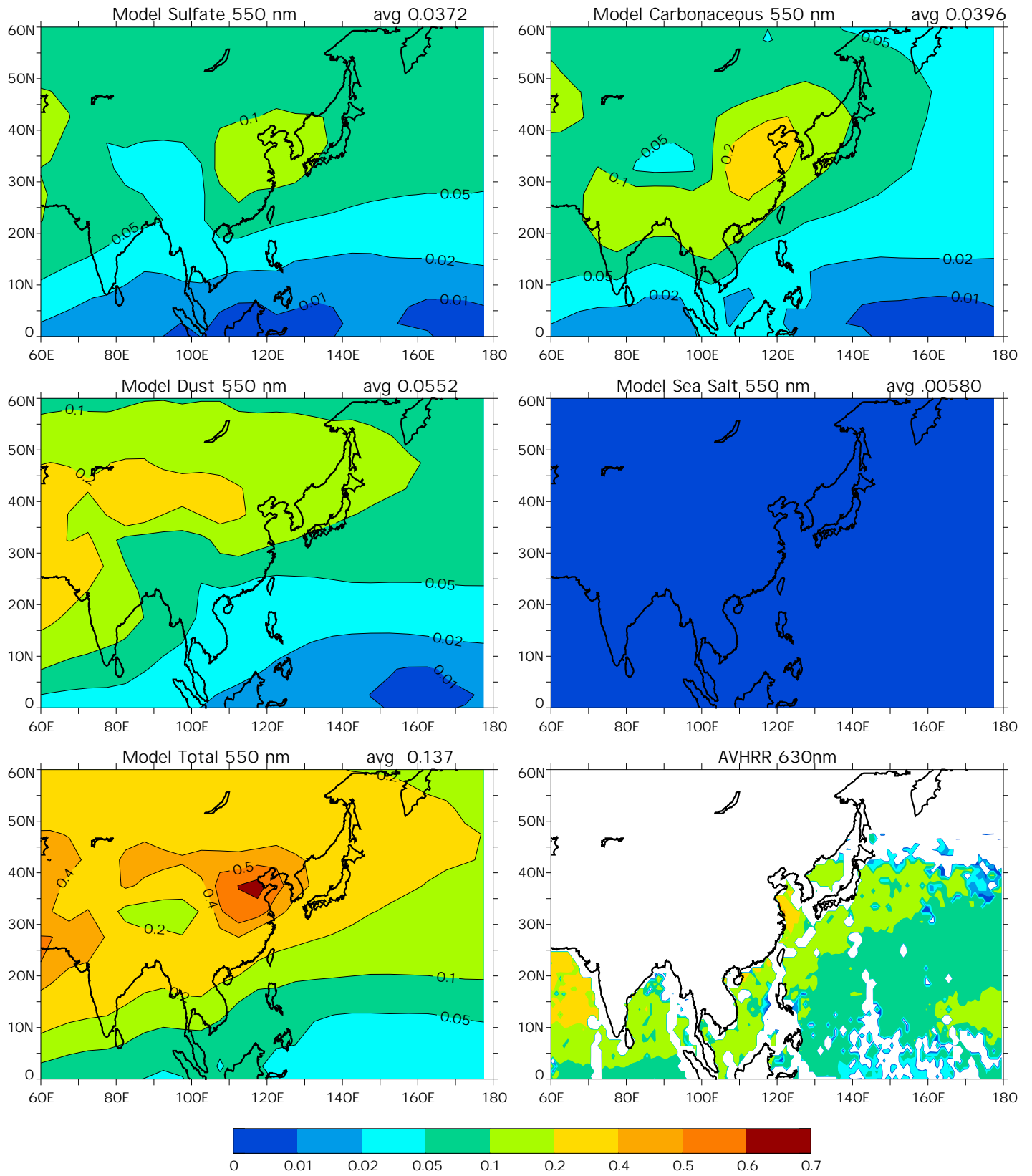


Figure 4. Aerosol optical thickness for 1990. Model results are for 550 nm (based on Chin et al, 1999; Ginoux et al., 2000; and unpublished results from Chin and Ginoux). AVHRR is for 630 nm.

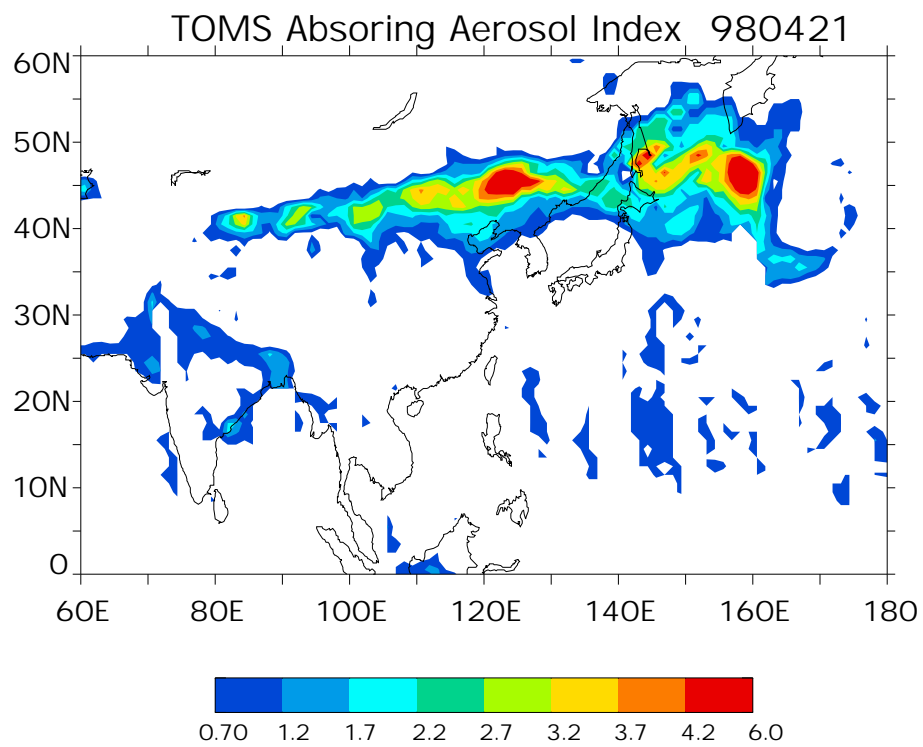


Figure 5. Asian dust storm and transport shown in the TOMS absorbing aerosol index